

# Two-site two-electron Holstein model: a perturbation study

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## Abstract

The two-site two-electron Holstein model is studied within a perturbation method based on a variational phonon basis obtained through the modified Lang-Firsov (MLF) transformation. The ground-state wave function and the energy are found out considering up to the seventh and eighth order of perturbation, respectively. The convergence of the perturbation corrections to the ground state energy as well as to the correlation functions are investigated. The kinetic energy and the correlation functions involving charge and lattice deformations are studied as a function of electron-phonon ( $e$ -ph) coupling and electron-electron interactions for different values of adiabaticity parameter. The simultaneous effect of the  $e$ -ph coupling and Coulomb repulsion on the kinetic energy shows interesting features.

## 1. Introduction

The Holstein model [1] is one of the fundamental models describing the interactions of conduction electrons with lattice vibrations and has been studied widely for a

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long time using different analytical and numerical methods. However, conventional analytical methods based on variational and perturbation approach are found to be much less satisfactory than the numerical methods; particularly in a wide region of intermediate values of electron-phonon ( $e$ -ph) coupling and hopping. The importance of a reliable analytical approach is crucial due to the failure of standard analytical approximations on the Holstein model; the Migdal approximation [2] is valid for a weakly coupled adiabatic system, while the Lang-Firsov canonical transformation [3] could not be suitable far from the nonadiabatic limit and strong coupling limit. The failure of the standard analytical techniques in the intermediate range of coupling has been observed by many workers [4] while comparing with exact results. Recently we [5] developed a perturbative expansion using different phonon bases obtained through the LF, MLF and MLF with squeezing (MLFS) transformations for a two-site single electron Holstein model. We found that the perturbation corrections within the MLF and MLFS methods are much smaller than the corresponding LF values for weak and intermediate couplings. The MLF method shows a very satisfactory convergence in perturbation expansion and is able to produce almost exact results for the entire region of the  $e$ -ph coupling from weak to intermediate values of hopping.

Our aim is to extend the applicability of our approach, developed for a single electron system, to a many-electron system. To examine that whether this perturbation method based on MLF phonon basis works well or not with increasing number of electrons and in presence of electron-electron ( $e$ - $e$ ) interaction, we consider a two-site two-electron Holstein model which is the minimal model to investigate the effect of  $e$ - $e$  interaction on the polaronic properties and the combined effects of the electronic correlation and the  $e$ -ph coupling on the kinetic energy of the system. It may be mentioned that the polaron crossover has been studied in two-site and four-site systems in presence of  $e$ - $e$  interaction by different groups [6, 7, 8] with the Holstein model previously. But most of the studies were done at the zero phonon approximation level with MLF or MLFS transformed Hamiltonian. However, the combined effect of  $e$ - $e$  interaction and  $e$ -ph interaction on the kinetic energy has not been properly studied and this is the main motivation of the present study.

In our study, we have developed a perturbation series using the MLF phonon basis and examined the convergence by comparing contributions of different orders of perturbation to the energy and the correlation functions for the ground state. The variation of different correlation functions involving charge and lattice deformation and the kinetic energy of the system with  $e$ -ph coupling are presented and discussed for different values of adiabaticity parameters and Coulomb repulsion.

## 2. Model Hamiltonian and formalism

The two-site two-electron Holstein Hamiltonian is

$$\begin{aligned}
H = & \sum_{i,\sigma} \epsilon n_{i\sigma} - \sum_{\sigma} t (c_{1\sigma}^{\dagger} c_{2\sigma} + c_{2\sigma}^{\dagger} c_{1\sigma}) + U \sum_i n_{i\uparrow} n_{i\downarrow} + V n_1 n_2 \\
& + g_1 \omega_0 \sum_{i,\sigma} n_{i\sigma} (b_i + b_i^{\dagger}) + g_2 \omega_0 \sum_{i,\sigma} n_{i\sigma} (b_{i+\delta} + b_{i+\delta}^{\dagger}) + \omega_0 \sum_i b_i^{\dagger} b_i
\end{aligned} \tag{1}$$

where  $i=1$  or  $2$ , denotes the sites.  $c_{i\sigma}$  ( $c_{i\sigma}^{\dagger}$ ) is the annihilation (creation) operator for the electron with spin  $\sigma$  at site  $i$  and  $n_{i\sigma}$  ( $=c_{i\sigma}^{\dagger} c_{i\sigma}$ ) is the corresponding number operator,  $g_1$  and  $g_2$  denote, respectively, the on-site and inter-site  $e$ -ph coupling strengths.  $t$  is the usual hopping integral.  $b_i$  ( $b_i^{\dagger}$ ) is the annihilation (creation) operator for the phonons corresponding to interatomic vibrations at site  $i$ ,  $\omega_0$  is the phonon frequency,  $U$  and  $V$  are, respectively, the on-site and inter-site Coulomb repulsion between the electrons.

Introducing new phonon operators  $\mathbf{a} = (b_1 + b_2)/\sqrt{2}$  and  $d = (b_1 - b_2)/\sqrt{2}$  the Hamiltonian is separated into two parts ( $H = H_d + H_{\mathbf{a}}$ ) [5]:

$$\begin{aligned}
H_d = & \sum_{i,\sigma} \epsilon n_{i,\sigma} - \sum_{\sigma} t (c_{1\sigma}^{\dagger} c_{2\sigma} + c_{2\sigma}^{\dagger} c_{1\sigma}) + U \sum_i n_{i\uparrow} n_{i\downarrow} + V n_1 n_2 \\
& + \omega_0 g_- (n_1 - n_2) (d + d^{\dagger}) + \omega_0 d^{\dagger} d
\end{aligned} \tag{2}$$

and

$$H_{\mathbf{a}} = \omega_0 \tilde{\mathbf{a}}^{\dagger} \tilde{\mathbf{a}} - \omega_0 n^2 g_+^2 \tag{3}$$

where  $g_+ = (g_1 + g_2)/\sqrt{2}$ ,  $g_- = (g_1 - g_2)/\sqrt{2}$ ,  $\tilde{\mathbf{a}} = \mathbf{a} + n g_+$ ,  $\tilde{\mathbf{a}}^{\dagger} = \mathbf{a}^{\dagger} + n g_+$ ,  $n = n_1 + n_2$  and  $n_i = n_{i\uparrow} + n_{i\downarrow}$ .

$H_{\mathbf{a}}$  describes a shifted oscillator which couples only with the total number of electrons  $n$ , which is a constant of motion.  $H_d$  represents an effective  $e$ -ph system where phonons directly couple with the electronic degrees of freedom. We now use the MLF transformation where the lattice deformations produced by the electron are treated as variational parameters [7, 9, 10]. For the present system the relevant transformation is,

$$\tilde{H}_d = e^R H_d e^{-R} \tag{4}$$

where  $R = \lambda(n_1 - n_2)(d^{\dagger} - d)$ ,  $\lambda$  is a variational parameter related to the displacement of the  $d$  oscillator.

The transformed Hamiltonian is then obtained as [7]

$$\begin{aligned}\tilde{H}_d = & \omega_0 d^\dagger d + \sum_{i,\sigma} \epsilon_p n_{i\sigma} - \sum_{\sigma} t [c_{1\sigma}^\dagger c_{2\sigma} \exp(2\lambda(d^\dagger - d)) + c_{2\sigma}^\dagger c_{1\sigma} \exp(-2\lambda(d^\dagger - d))] \\ & + \omega_0(g_- - \lambda)(n_1 - n_2)(d + d^\dagger) + U_e \sum_i n_{i\uparrow} n_{i\downarrow} + V_e n_1 n_2\end{aligned}\quad (5)$$

where

$$\epsilon_p = \epsilon - \omega_0(2g_- - \lambda) \lambda \quad (6)$$

$$U_e = U - 2\omega_0(2g_- - \lambda) \lambda \quad (7)$$

$$V_e = V + 2\omega_0(2g_- - \lambda) \lambda \quad (8)$$

For the two-site two-electron system there are six electronic states of which three belong to the triplet states as follows:

$$|\psi_{T1}\rangle = c_{1\uparrow}^\dagger c_{2\uparrow}^\dagger |0\rangle_e \quad (9)$$

$$|\psi_{T2}\rangle = \frac{1}{\sqrt{2}} (c_{1\uparrow}^\dagger c_{2\downarrow}^\dagger + c_{1\downarrow}^\dagger c_{2\uparrow}^\dagger) |0\rangle_e \quad (10)$$

$$|\psi_{T3}\rangle = c_{1\downarrow}^\dagger c_{2\downarrow}^\dagger |0\rangle_e \quad (11)$$

and other three states are singlet as given below:

$$|\psi_1\rangle = \frac{1}{\sqrt{2}} (c_{1\uparrow}^\dagger c_{1\downarrow}^\dagger - c_{2\uparrow}^\dagger c_{2\downarrow}^\dagger) |0\rangle_e \quad (12)$$

$$|\psi_2\rangle = \frac{1}{\sqrt{2}} (c_{1\uparrow}^\dagger c_{1\downarrow}^\dagger + c_{2\uparrow}^\dagger c_{2\downarrow}^\dagger) |0\rangle_e \quad (13)$$

$$|\psi_3\rangle = \frac{1}{\sqrt{2}} (c_{1\uparrow}^\dagger c_{2\downarrow}^\dagger - c_{1\downarrow}^\dagger c_{2\uparrow}^\dagger) |0\rangle_e \quad (14)$$

The triplet states do not mix with each other or couple with the  $d$ -oscillator. In the following we will only consider the singlet states and the  $d$ -oscillator which are mutually coupled through the off-diagonal terms of the Hamiltonian (5). For a perturbation method it is desirable to use a basis where the major part of the Hamiltonian becomes diagonal. For this purpose we choose the basis set  $|\psi_+, N\rangle$ ,  $|\psi_1, N\rangle$  and  $|\psi_-, N\rangle$  defined as

$$|\psi_+, N\rangle = \frac{1}{\sqrt{a_N^2 + b_N^2}} (a_N |\psi_2\rangle + b_N |\psi_3\rangle) |N\rangle \quad (15)$$

$$|\psi_1, N\rangle = |\psi_1\rangle|N\rangle \quad (16)$$

$$|\psi_-, N\rangle = \frac{1}{\sqrt{a_N^2 + b_N^2}}(a_N|\psi_3\rangle - b_N|\psi_2\rangle)|N\rangle \quad (17)$$

where  $|\psi_+\rangle$ ,  $|\psi_1\rangle$  and  $|\psi_-\rangle$  represent the bonding, nonbonding and antibonding electronic states, respectively and  $|N\rangle$  denotes the  $N$ th excited oscillator state in the MLF phonon basis where  $N$  takes integer values from 0 to  $\infty$ .

Here  $a_N = -(U_e - V_e) + \sqrt{(U_e - V_e)^2 + 16t_e^2(N, N)}$  and  $b_N = 4t_e(N, N)$ .

where  $t_e(N, N) = t_e \sum_{p=1}^N (-1)^p \frac{(2\lambda)^{2p}}{p!} N_{C_p}$ ,  $N_{C_p} = \frac{N!}{p!(N-p)!}$ , and  $t_e = t \exp(-2\lambda^2)$ .

The hopping term in Eq. (5) has both diagonal and offdiagonal matrix elements in the chosen basis (Eqs. 15-17). The states  $|\psi_e, N\rangle$  (for  $e = +, 1, -$ ) are the eigenstates of the unperturbed Hamiltonian

$$\begin{aligned} \tilde{H}_0 &= \omega_0 d^\dagger d + \sum_i \epsilon_p n_i - \sum_\sigma t_e F(d^\dagger d) [c_{1\sigma}^\dagger c_{2\sigma} + c_{2\sigma}^\dagger c_{1\sigma}] \\ &+ U_e \sum_i n_{i\uparrow} n_{i\downarrow} + V_e n_1 n_2 \end{aligned} \quad (18)$$

It may be noted that  $t_e F(d^\dagger d)$  in the above Eq. (18) is the diagonal part of the operator  $t \exp[\pm 2\lambda(d_i^\dagger - d_i)]$  in Eq. (5) in the phonon occupation number representation.

$$\begin{aligned} \text{Since } \exp[2\lambda(d^\dagger - d)] &= \exp[2\lambda d^\dagger] \exp[-2\lambda d] \exp\left[\frac{4\lambda^2}{2}[d^\dagger, d]_-\right] \\ &= e^{-2\lambda^2} \sum_p \sum_q (-1)^q \frac{(2\lambda)^{p+q}}{p! q!} (d^\dagger)^p (d)^q, \\ \text{so } F(d^\dagger d) &= \sum_{p=1}^N (-1)^p (2\lambda)^{2p} (d^\dagger d)^p / (p!)^2. \end{aligned}$$

For the choice of the unperturbed Hamiltonian in Eq. (18) the perturbation Hamiltonian ( $\tilde{H} - \tilde{H}_0$ ) has no diagonal matrix element in our chosen basis. This is not achieved when one replaces  $F(d^\dagger d)$  by 1 and  $a_N$ ,  $b_N$  become  $N$ -independent. The convergence in perturbation expansion for the latter case is found to be weaker than that when  $F(d^\dagger d)$  is included in  $H_0$ .

The unperturbed energies corresponding to the eigenstates  $|\psi_e, N\rangle$  ( $e = +, 1, -$ ) are

$$E_{+,N}^{(0)} = N\omega_0 + 2\epsilon_p + \frac{(U_e + V_e)}{2} - \frac{1}{2} \sqrt{(U_e - V_e)^2 + 16t_e^2(N, N)},$$

$$\begin{aligned}
E_{1,N}^0 &= 2\epsilon_p + U_e \\
\text{and } E_{-,N}^{(0)} &= N\omega_0 + 2\epsilon_p + \frac{(U_e + V_e)}{2} + \frac{1}{2}\sqrt{(U_e - V_e)^2 + 16t_e^2(N, N)} \quad (19)
\end{aligned}$$

The part of the Hamiltonian, which is not included in  $\tilde{H}_0$ , is taken as the perturbation  $\tilde{H}_1$ . For the chosen basis, the state  $|\psi_+, 0\rangle$  has the lowest unperturbed energy  $E_{+,0}^{(0)}$ . The general off-diagonal matrix elements of  $\tilde{H}_1$  between the states  $|\psi_\pm, N\rangle$  and  $|\psi_1, M\rangle$  are calculated as (for  $(N - M) > 0$ )

$$\begin{aligned}
\langle N, \psi_\pm | \tilde{H}_1 | \psi_\pm, M \rangle &= \mp 2t_e(N, M) (a'_N b'_M + a'_M b'_N) \quad \text{for even } (N - M). \\
&= 0 \quad \text{for odd } (N - M). \quad (20)
\end{aligned}$$

$$\begin{aligned}
\langle N, \psi_\pm | \tilde{H}_1 | \psi_\mp, M \rangle &= -2t_e(N, M) (a'_N a'_M - b'_M b'_N) \quad \text{for even } (N - M). \\
&= 0 \quad \text{for odd } (N - M). \quad (21)
\end{aligned}$$

$$\begin{aligned}
\langle N, \psi_1 | \tilde{H}_1 | \psi_+, M \rangle &= 2a'_M P(N, M) - 2t_e(N, M) b'_M \quad \text{for odd } (N - M). \\
&= 0 \quad \text{for even } (N - M). \quad (22)
\end{aligned}$$

$$\begin{aligned}
\langle N, \psi_1 | \tilde{H}_1 | \psi_-, M \rangle &= -2b'_M P(N, M) - 2t_e(N, M) a'_M \quad \text{for odd } (N - M). \\
&= 0 \quad \text{for even } (N - M). \quad (23)
\end{aligned}$$

$$\begin{aligned}
\text{where } t(N, M) &= t_e(2\lambda)^{N-M} \sqrt{\frac{N!}{M!}} \left[ \frac{1}{(N-M)!} + \sum_{R=1}^M [(-1)^R \right. \\
&\quad \left. \frac{(2\lambda)^{2R}}{(N-M+R)!R!} M(M-1)\dots(M-R+1) \right] \\
P(N, M) &= \omega_0(g_- - \lambda)[\sqrt{M} \delta_{N,M-1} + \sqrt{M+1} \delta_{N,M+1}] \\
a'_N &= \frac{a_N}{\sqrt{(a_N^2 + b_N^2)}} \quad \text{and} \quad b'_N = \frac{b_N}{\sqrt{(a_N^2 + b_N^2)}}
\end{aligned}$$

Now one has to make a proper choice of  $\lambda$  so that the perturbative expansion is satisfactorily convergent. In our previous work we obtained good convergence of the perturbation series for the value of  $\lambda$  which minimizes the unperturbed ground state energy. Following the same procedure, i.e. minimizing  $E_{+,0}^{(0)}$  with respect to  $\lambda$  we obtain

$$\lambda = g_- / \left[ 1 + \frac{4t_e}{\omega_0[\sqrt{r^2 + 4} - r]} \right] \quad (24)$$

where  $r = (U_e - V_e)/2t_e$ .

The corrected ground state wave function may be written as,

$$|\psi_G\rangle = \frac{1}{\sqrt{N_G}} \left[ |\psi_+, 0\rangle + \sum_{N=1,3,..} C_N^1 |\psi_1, N\rangle + \sum_{N=2,4,..} C_N^\pm |\psi_\pm, N\rangle \right] \quad (25)$$

where  $N_G = 1 + \sum_{N=1,3,..} (C_N^1)^2 + \sum_{N=2,4,..} ((C_N^+)^2 + (C_N^-)^2)$  is the normalization factor.

The coefficients  $C_N^1$ ,  $C_N^+$  and  $C_N^-$  for different values of  $N$  are determined using the matrix elements of  $\tilde{H}_1$  connecting different unperturbed states and their energies following Ref. [5]. These Coefficients are determined up to the seventh order of perturbation while the ground-state energy is found out up to the eighth order of perturbation.

We have also studied the static correlation functions involving charge and lattice deformations  $\langle n_1 u_1 \rangle_0$  and  $\langle n_1 u_2 \rangle_0$ , where  $u_1$  and  $u_2$  represents the lattice deformations at sites 1 and 2 respectively. These correlation functions may be written in terms of relevant operators as

$$n_1 u_{1,2} = n_1 \left[ -2g_+ + \frac{(\mathbf{a} + \mathbf{a}^\dagger)}{2} \right] \pm n_1 \left[ -\lambda(n_1 - n_2) + \frac{(d + d^\dagger)}{2} \right] \quad (26)$$

The final form of the correlation functions are obtained as

$$\begin{aligned} \langle n_1 u_{1,2} \rangle_0 &= -2g_+ \mp \frac{2\lambda}{N_G} \left[ \frac{a_0^2}{a_0^2 + b_0^2} + \sum_{N=1,3,5,..} (C_N^1)^2 \right. \\ &+ \sum_{N=2,4,..} \frac{1}{a_N^2 + b_N^2} (a_N C_N^+ - b_N C_N^-)^2 \left. \right] \pm \frac{1}{N_G} \left[ \frac{a_0}{\sqrt{a_0^2 + b_0^2}} C_1^1 \right. \\ &+ \sum_{N=2,4,..} \frac{\sqrt{N} C_{N-1}^1}{\sqrt{a_N^2 + b_N^2}} (a_N C_N^+ - b_N C_N^-) \\ &+ \sum_{N=3,5,..} \frac{\sqrt{N} C_N^1}{\sqrt{a_N^2 + b_N^2}} (a_{N-1} C_{N-1}^+ - b_{N-1} C_{N-1}^-) \left. \right] \end{aligned} \quad (27)$$

The kinetic energy of the system in the ground state  $E_{ke} = -t < \psi_G | \sum_\sigma [c_{1\sigma}^\dagger c_{2\sigma} \exp(2\lambda(d^\dagger - d)) + c_{2\sigma}^\dagger c_{1\sigma} \exp(-2\lambda(d^\dagger - d))] | \psi_G \rangle$ , has also been calculated using the ground state wave function  $|\psi_G\rangle$ , evaluated up to the seventh order of perturbation.

### 3. Results and discussions

From the relevant analytical expressions, the quantities of our interest have been calculated within the LF and the MLF perturbation methods considering 35 phonon states (which is more than sufficient for  $g_- \leq 2.5$  in the transformed phonon basis). For all numerical calculations we take  $\epsilon = 0$ ,  $g_2 = 0$  (i.e.,  $g_+ = g_-$ ) in this work.

In figure 1 we have shown the relative perturbation corrections to the ground-state energy, i.e., the ratios of the perturbation corrections of different orders to the unperturbed ground-state energy as a function of  $g_-$  for  $t/\omega_0 = 1$ . It appears from figure 1 that the convergence is more or less satisfactory in both the MLF and LF approaches. Apart from the second order correction, higher order energy corrections are very small within the MLF approach except in a small region of  $g_- \sim 0.6$  to  $0.8$  where the convergence is relatively weaker. The energy corrections of fifth to eighth order are, however, negligibly small even in this region. The energy correction of any order within the LF method is larger than the corresponding correction within the MLF approach. For smaller values of  $t/\omega_0$  the corrections are much less and the convergence is better as expected from our previous study [5].

To examine the applicability of our method for higher values of  $t$  and also in presence of  $e$ - $e$  interaction, we plot in figure 2 the relative perturbation corrections of different orders to the ground-state energy as a function of  $g_-$  for  $t/\omega_0 = 2$  for both zero and nonzero values of  $e$ - $e$  interactions. Figure 2(a) shows that the convergence is satisfactory and except the second order correction, higher order energy corrections are small. In presence of electronic interactions (nonzero  $U$  and  $V$ ), magnitudes of the relative energy corrections increase but the convergence is still good (figure 2(b)). We calculate the ground-state energy within the MLF method up to the eighth order of perturbation. As pointed out previously, the fifth to eighth order energy corrections are so small that the energy calculated up to the eighth order of perturbation may be treated as an almost exact result. We find that our results exactly match with that obtained by the exact diagonalization study [11] for the same set of parameters.

The on-site and inter-site charge-lattice deformation correlations,  $\langle n_1 u_1 \rangle_0$  and  $\langle n_1 u_2 \rangle_0$ , are determined for the ground state considering up to the seventh order of perturbation within the MLF method. In figure 3 we have plotted these quantities



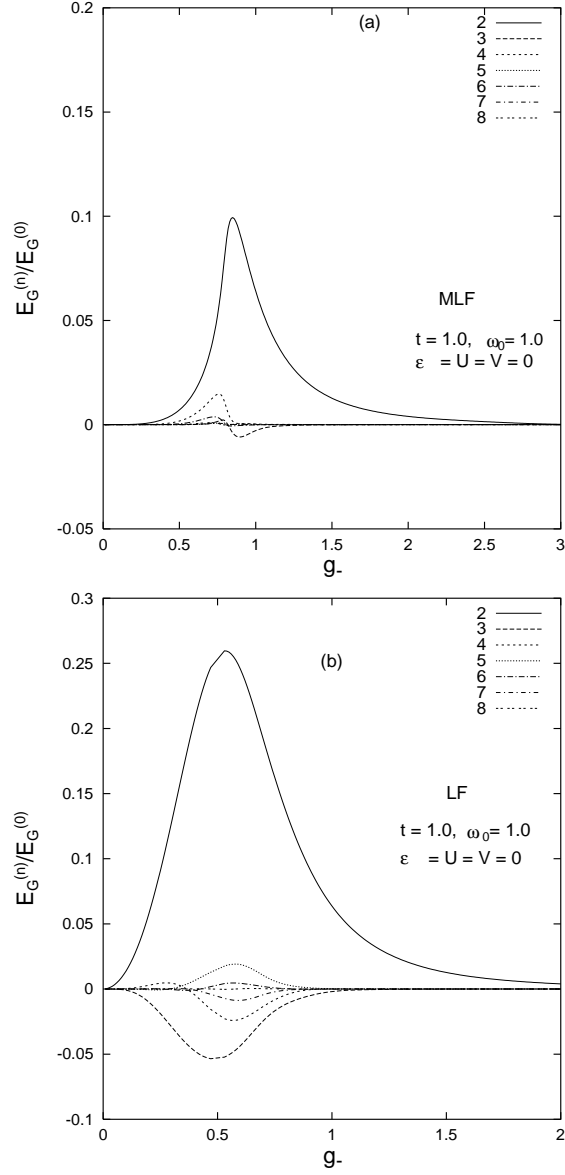


Figure 1: Variation of the relative perturbation corrections  $E_G^{(n)}/E_G^{(0)}$  to the ground state energy as a function of the coupling strength ( $g_-$ ) for  $t/\omega_0 = 1.0$  for (a) MLF method and (b) LF method.  $E_G^{(n)}$  is the  $n$ th order perturbation correction to the ground state energy and  $E_G^{(0)} = E_{+,0}^{(0)}$  is the unperturbed ground state energy. 2,...,8 denote the value of  $n$  in  $E_G^{(n)}$ .

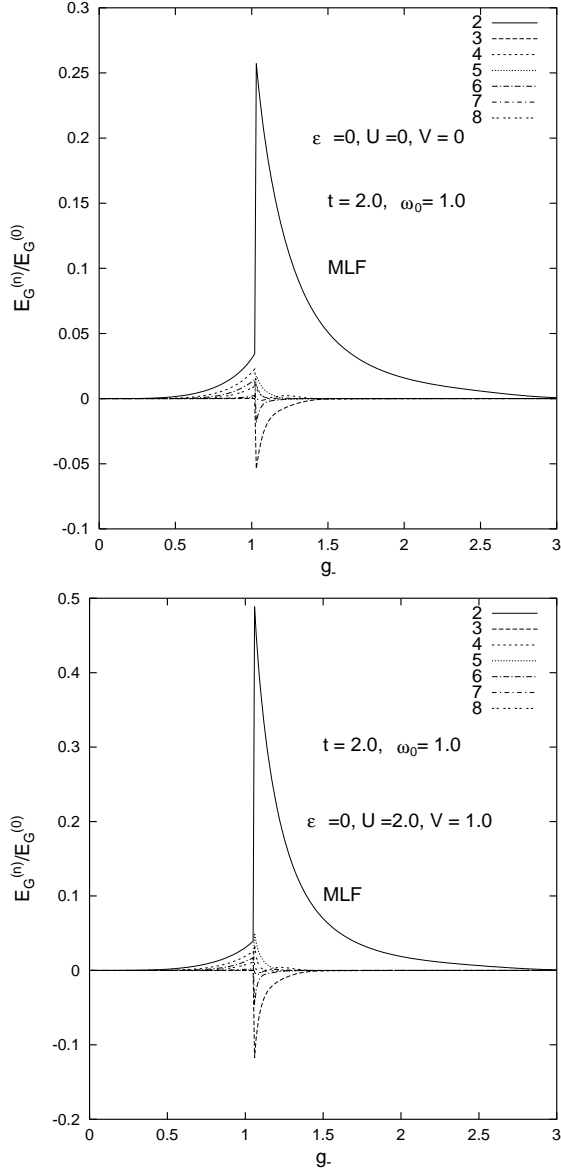


Figure 2: Variation of the relative perturbation corrections  $E_G^{(n)}/E_G^{(0)}$  to the ground state energy as a function of the coupling strength ( $g_-$ ) for  $t/\omega_0 = 2.0$  for the MLF method for (a)  $U = 0$  and (b)  $U = 2.0, V = 1.0$ .  $E_G^{(n)}$  is the  $n$ th order perturbation correction to the ground state energy and  $E_G^{(0)} = E_{+,0}^{(0)}$  is the unperturbed ground state energy. 2,...,8 denote the value of  $n$  in  $E_G^{(n)}$ .

against  $g_-$ . When we examine the convergence by comparing the values of the same correlation function, calculated up to different orders of perturbation we find that the values obtained considering up to the fourth to the seventh order of perturbation, remain almost same. This ensures a good convergence of our perturbation method for the correlation function and thus the results obtained are expected to match satisfactorily with the exact result. The unperturbed LF and the MLF results are also plotted in figure 3 for a comparison, which shows that the unperturbed MLF results for the correlation functions are much closer to the exact result than the corresponding LF result. The relative difference between the exact MLF result and the MLF (ZPA) result is much less for  $\langle n_1 u_1 \rangle_0$  than for  $\langle n_1 u_2 \rangle_0$ . This is owing to the fact that  $\langle n_1 u_2 \rangle_0$  is a very sensitive function of the perturbation corrections to the wave function compared to  $\langle n_1 u_1 \rangle_0$ . The MLF perturbation result predicts a long tail for  $\langle n_1 u_2 \rangle_0$  in the strongly coupled region where both the LF (ZPA) and the MLF (ZPA) give zero value for  $\langle n_1 u_2 \rangle_0$ . This result is a signature of the retardation effect even in the strong coupling region where even the MLF (ZPA) do not predict the retardation effect.

To show the crossover from the delocalized (large) to localized (small) polaron we plot the quantity  $\chi_{12} (= \langle (n_1 - n_2)(u_1 - u_2) \rangle_0 / g_+)$  against  $g_-$  in figure 4(a) where the MLF perturbation results for different values of  $t/\omega_0$  are shown. The ZPA results within MLF and LF methods are also given for  $t/\omega_0 = 1.0$  to compare them with the exact perturbation results. The MLF (ZPA) result is very close to the MLF perturbation result. The MLF perturbation result shows a smooth crossover from large to small polaron as  $e$ -ph coupling strength increases. If we identify the crossover point as the point of inflection of the curve obtained within the MLF (perturbation) approach we find that for  $t/\omega_0 = 1$  the crossover occurs at a critical  $e$ -ph coupling  $g_-$  ( $g_- \sim 0.8$ ) when  $U = V = 0$ . At this point the coefficient of one phonon state ( $C_{N=1}^1$  in Eq. (25)) changes its sign. Similar behaviour has been also observed in case of our previous single polaron studies [5]. The critical value of the  $e$ -ph coupling where polaron crossover occurs increases with increasing value of  $t/\omega_0$ , as expected.

In figure 4(b) we have shown the effect of  $e$ - $e$  interaction on the polaron crossover by plotting  $\chi_{12}$  against  $g_-$  for several values of  $U$  and  $V$ . For this two-site two-electron system the polaronic properties we have studied, depend on  $(U - V)$  rather than  $U$  and  $V$  independently. From figure 4(b) it is seen that the critical coupling, where the

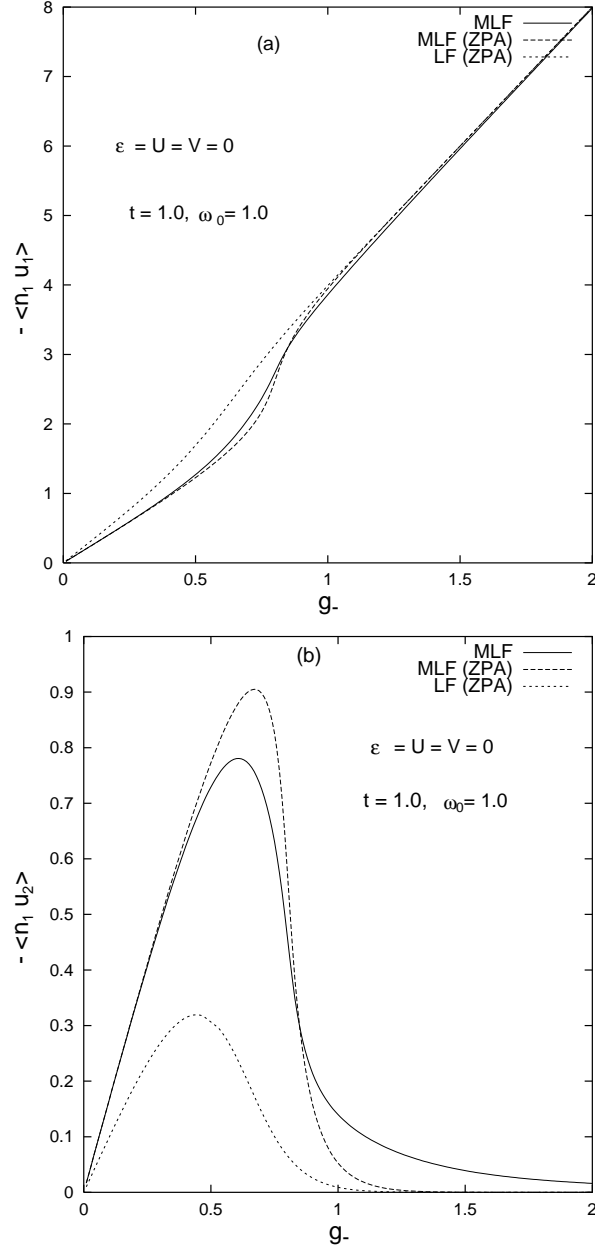


Figure 3: Plot of the correlation functions (a)  $\langle n_1 u_1 \rangle_0$  and (b)  $\langle n_1 u_2 \rangle_0$  with  $g_-$  for  $t/\omega_0 = 1.0$  along with the ZPA results within LF and MLF methods. The solid curve corresponds to that obtained within the MLF method considering up to seventh order correction to the ground-state wave function.

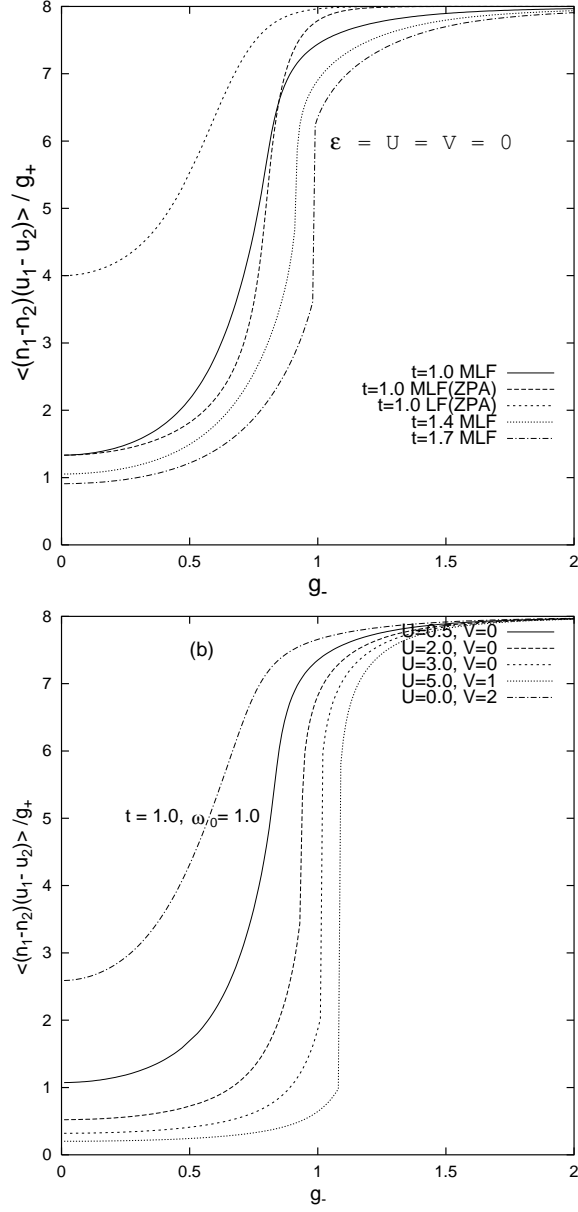


Figure 4: The variations of  $\chi_{12} = \langle (n_1 - n_2)(u_1 - u_2) \rangle_0 / g_+$  with  $g_-$  within MLF perturbation method (a) for different values of  $t/\omega_0$  and  $U = 0$ ; the MLF (ZPA) and LF (ZPA) results for  $t/\omega_0 = 1.0$  are also shown. (b) for different values of  $U$  and  $V$  for  $t/\omega_0 = 1.0$ .

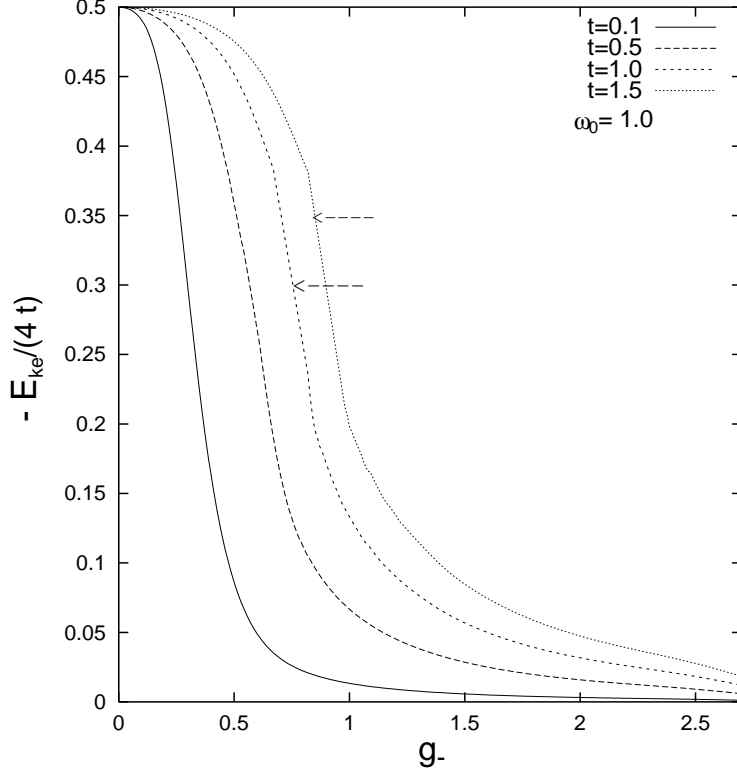


Figure 5: The kinetic energy of the system, obtained using the seventh order of perturbation within MLF method, as a function of  $g_-$  for different values  $t/\omega_0$  ( $=0.1, 0.5, 1.0$  and  $1.5$ ).

polaron crossover takes place, increases and the crossover becomes more abrupt with increasing  $(U - V)$ . Increasing on-site Coulomb repulsion makes the crossover region more abrupt and extends the large polaron region. Similar results were also reported earlier [6]. The inter-site Coulomb repulsion, on the other hand, makes the crossover smoother and the crossover takes place at a lower value of  $e$ -ph coupling.

The variation of the kinetic energy of the system with  $e$ -ph coupling strength  $g_-$  is shown in figure 5 for  $U = V = 0$  and for different values of  $t/\omega_0$  ( $= 0.1, 0.5, 1.0$

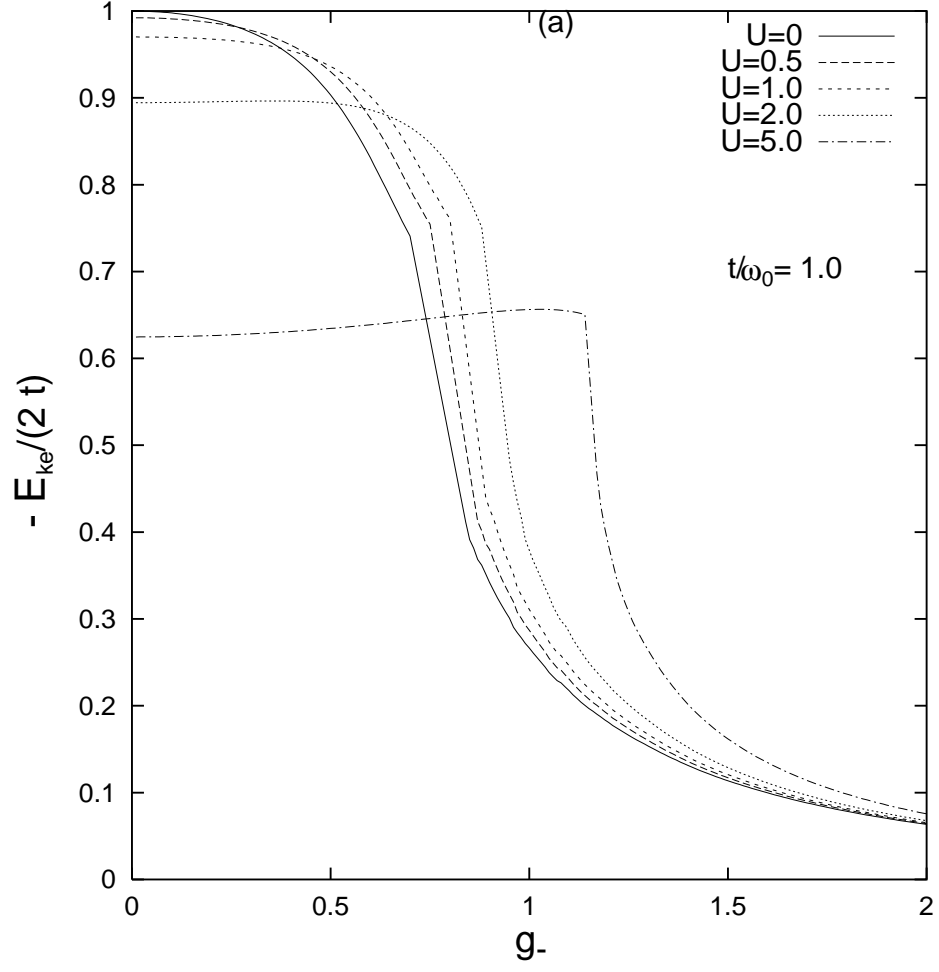


Figure 6: For  $t/\omega_0 = 1.0$ , the kinetic energy of the system, obtained by the MLF perturbation method, as a function of  $g_-$  for different values of  $U$ .

and 1.5). We find that our perturbation results for the kinetic energy match exactly with the results obtained by exact diagonalization study [11] for the corresponding set of parameters. It may be mentioned that for  $t/\omega_0 = 1.0, 1.5$  the convergence is not good in a very small region of  $g_-$  around the points indicated by the arrows in the figure. Except these narrow regions the convergence of our perturbation method is excellent in the rest of the parameter space we studied.

It is well known that the kinetic energy of a tight-binding electronic system is suppressed by the introduction of either  $e$ -ph coupling or by Coulomb repulsion. An important question one may ask what is the simultaneous role of the Coulomb repulsion and the  $e$ -ph interaction in lowering the kinetic energy of the system. To address this question we have plotted the kinetic energy against  $e$ -ph coupling strength for different values of  $U$  for  $t/\omega_0 = 1.0$  in figure 6. It is seen from figure 6 that for large  $U$  values there is a flat region where the kinetic energy remains unaffected by increase of  $e$ -ph coupling. Higher the value of  $U$  larger is the flat region. Beyond this flat region the kinetic energy is suppressed almost exponentially with increasing  $g_-$ . The occurrence of this flat region and then a rapid fall in the kinetic energy for higher values of  $U$  may be understood from the results of figure 4(b) where it is found that for larger values of  $U$  the polaron crossover is abrupt but there is an extended large polaron region (in  $g_-$  space). The value of  $\chi_{12}$ , which is a measure of the difference in the lattice distortions produced at the charge residing site and the neighbouring site, is small and almost constant in this large polaron region. As a result a flat behaviour in the kinetic energy is obtained. Beyond a critical value of  $g_-$  an abrupt increase in  $\chi_{12}$  would lead to a sharp fall in the kinetic energy, which is evident in figure 6.

In figure 7 the variation of the kinetic energy with  $U$  for different values of  $g_-$  is shown. It is found that for  $g_- = 1.0$  and  $1.5$  there is a region where the kinetic energy increases with  $U$  in contrast to the usual behaviour that the kinetic energy is suppressed by the Coulomb correlation. The latter behaviour is of course seen in major parameter space. To understand this unusual feature of increasing kinetic energy with  $U$  for finite  $g_-$ , we plot the variation of  $\chi_{12}$  as well as the kinetic energy with  $U$  in the inset of figure 7 for  $g_- = 1.0$ . It is readily seen that as  $U$  increases the correlation function  $\chi_{12}$  decreases which results in an increase in the kinetic energy. At the crossover region,  $\chi_{12}$  shows a sharp fall which is reflected by the abrupt rise in the kinetic energy. For large values of  $U$ ,  $\chi_{12}$  is small and the polaron is a large



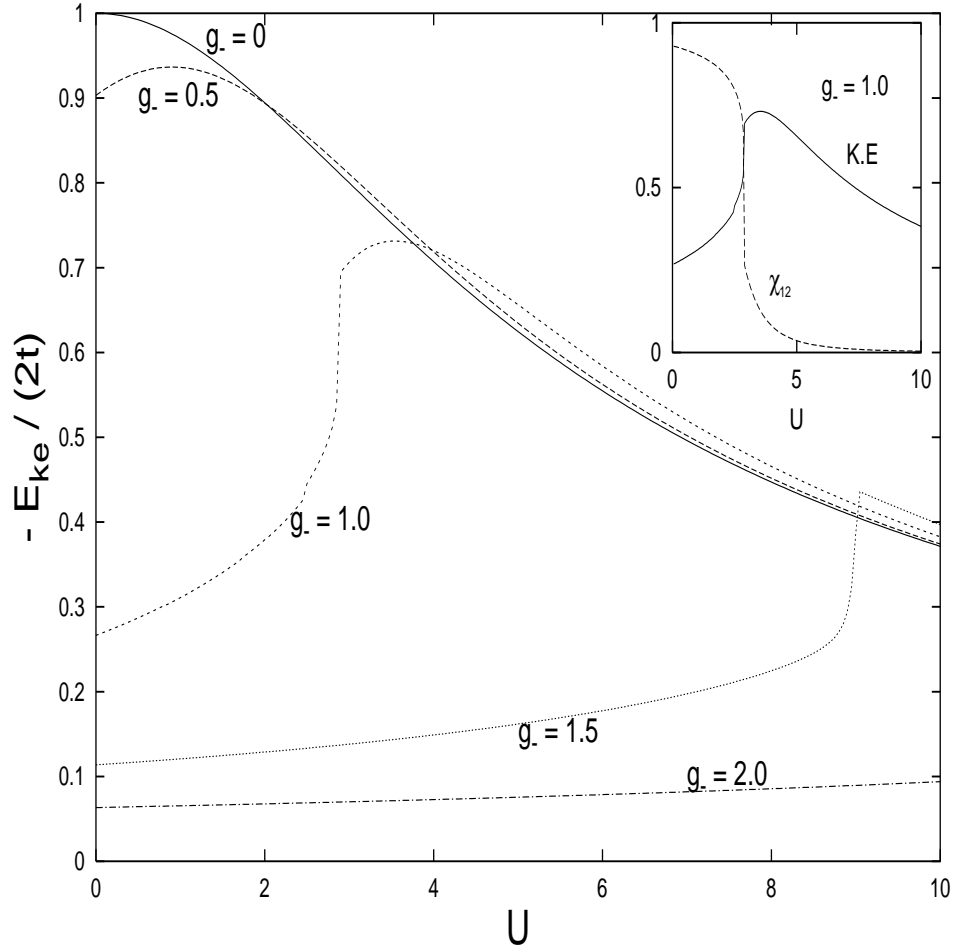


Figure 7: The kinetic energy of the system, obtained by the MLF perturbation method, as a function of  $U$  for different values of  $g_-$  for  $t/\omega_0 = 1.0$ ; The inset figure shows the variation of  $\chi_{12}/8$  with  $U$  for  $g_- = 1.0$ .

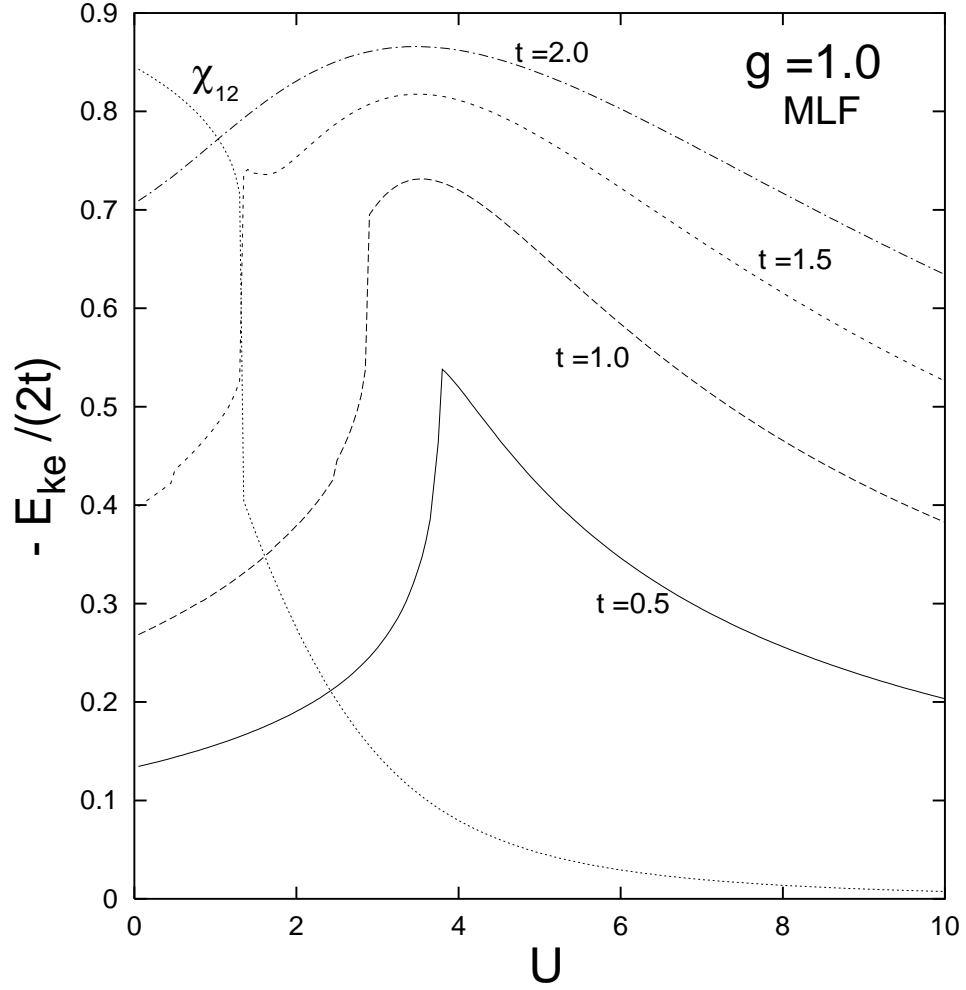


Figure 8: The kinetic energy of the system, obtained by the MLF perturbation method, as a function of  $U$  for a particular value of  $e$ -ph coupling ( $g_- = 1.0$ ) for different values of  $t/\omega_0$  ( $=0.5, 1.0, 1.5$  and  $2.0$ ).  $\chi_{12}/8$  is also plotted for  $t/\omega_0 = 1.5$ .

one. In this case the lattice distortions uniformly spread throughout the whole lattice (hence the charge particle behaves more like a normal electron) and the kinetic energy is suppressed with increasing  $U$  as commonly expected.

In figure 8 the variation of the kinetic energy with  $U$  for different values of  $t/\omega_0$  is shown for a fixed value of  $e$ -ph coupling ( $g_- = 1.0$ ). The kinetic energy increases with  $U$ , shows a peak and then decreases. Similar behaviour is also observed in figure 7. The peak is broader for higher values of  $t$ . The initial enhancement of the kinetic energy with  $U$  is due to rapid spread up of the polaron size or decrease in the value of  $\chi_{12}$  whereas in the large polaron region  $U$  plays its conventional role of suppressing the kinetic energy. With increasing  $t$  the kinetic energy increases as well as the large polaron region is extended. We have also plotted  $\chi_{12}$  in the same figure for  $t/\omega_0 = 1.5$  to show the rise in the kinetic energy is related to rapid fall in  $\chi_{12}$ . For  $t/\omega_0 = 2.0$ , the polaron is a large one for  $g_- = 1$  and so the small polaron region is not seen for this case.

In summary we apply the MLF perturbation method on a two-site two-electron Holstein model taking the diagonal part of the Hamiltonian (in the momentum space of the MLF basis) as the unperturbed Hamiltonian. A good convergence is achieved by our approach. The ground state energy and the kinetic energy obtained within this method match exactly with those obtained by exact diagonalization study. The behaviour of different correlation functions involving charge and the lattice distortions are reported as a function of  $e$ -ph coupling. Effect of the Coulomb repulsion and the adiabaticity parameter on the large to small polaron crossover is studied. On-site Coulomb repulsion makes the polaron crossover more abrupt. Our study on the kinetic energy in presence of both  $e$ -ph interaction and Coulomb repulsion shows that for large values of  $U$ , the kinetic energy is not suppressed by  $e$ -ph coupling in a range which is wider larger the value of  $U$ . In this range the polaron is a large one with almost an uniform value of  $\chi_{12}$ . This makes the kinetic energy insensitive to the  $e$ -ph coupling. When  $e$ -ph coupling crosses a critical value a rapid crossover to a small polaron occurs and in this region the  $e$ -ph coupling is effective in suppressing the kinetic energy. On the other hand for finite  $e$ -ph coupling the kinetic energy increases initially with  $U$  and then decays. This is ascribed to the fact that the former region corresponds to a small polaron or a crossover region where the main role of  $U$  is to spread the size of the polaron and make it more delocalized. This results in an increase in the kinetic energy whereas for large  $U$  values the polaron is a large one and the on-site correlation suppresses the kinetic energy in its usual way.

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